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Catalytic Treatment of Air Stripping Effluents

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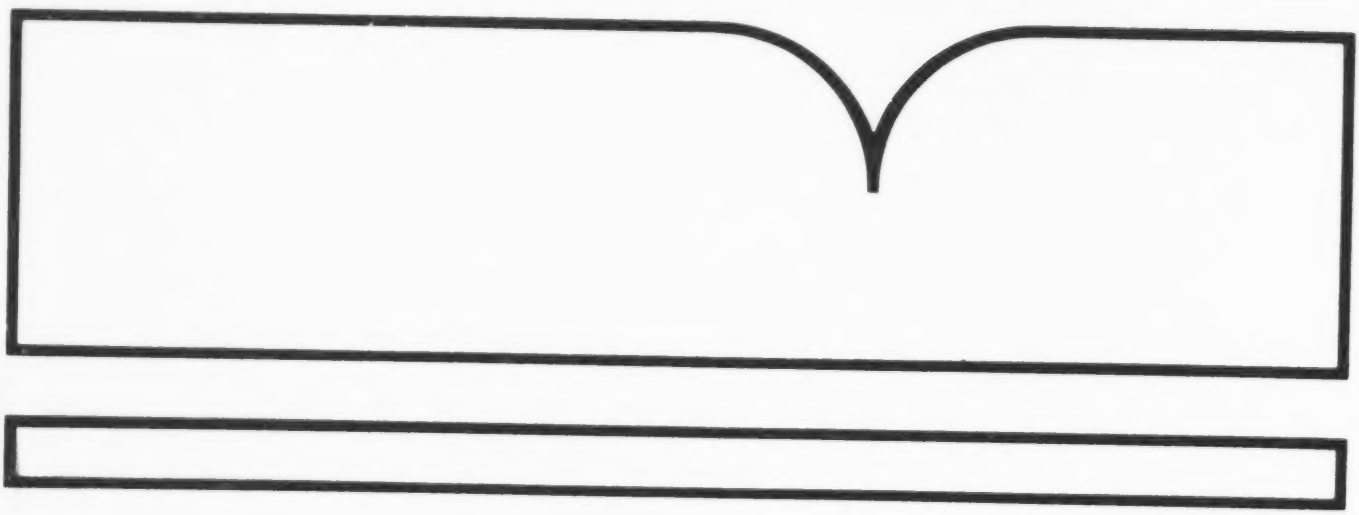

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CATALYTIC TREATMENT OF AIR STRIPPING EFFLUENTS

By

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16. ABSTRACT The paper reviews the applicability of catalytic oxidation to control groundwater air stripping gaseous effluents with emphasis on system designs and case histories. The contaminants and catalyst poisons encountered in stripping operations are also reviewed. Air stripping is an efficient and cost effective way to remove volatile organic contaminants from groundwater and soil. The removal of dissolved fuel fractions and chlorinated solvents at efficiencies in excess of 95% has been demonstrated on several occasions. Unfortunately, these contaminants are transferred to the air where they may continue to pose an environmental and health threat. Regulation of such emissions has already been considered in many states, including Michigan and California. However, few options are available for their control. Vapor-phase carbon adsorption and thermal incineration, the treatment methods which have been applied most often, have some disadvantages: adsorption merely transfers the contaminant to a solid phase, which in turn requires disposal or regeneration; and thermal incineration may be expensive, since it requires a substantial energy input to destroy dilute gas-phase contaminants. A new alternative is appearing in the form of catalytic oxidation. Like thermal incineration, it is an ultimate disposal method, but the energy costs are lower.

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Catalysis	Stationary Sources	07D 20M
Oxidation	Air Stripping	07B, 07C
Stripping (Distillation)	Catalytic Oxidation	07A, 13H
Effluents		
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Abstract

Air stripping has been shown to be an efficient and cost effective method of removing volatile organic contaminants from groundwater and soil. The removal of dissolved fuel fractions and chlorinated solvents at efficiencies in excess of 95% has been demonstrated on several occasions. Unfortunately, these contaminants are transferred to the air where they may continue to pose an environmental and health threat. Regulation of such emissions has already been considered in many states including Michigan and California. However, few options are available for their control. Vapor phase carbon adsorption and thermal incineration are the two treatment methods which have been applied most often. These technologies have some disadvantages. Adsorption merely transfers the contaminant to a solid phase, which in turn requires disposal or regeneration. Thermal incineration may be expensive, since it requires a substantial energy input to destroy dilute gas phase contaminants. A new alternative is appearing in the form of catalytic oxidation. Like thermal incineration, it is an ultimate disposal method, but since it operates at much lower temperatures, the energy costs are also lower. This paper will review the applicability of catalytic oxidation to control groundwater air stripping gaseous effluents with special attention given to system designs and case histories. The variety of contaminants and catalyst poisons encountered in stripping operations are also reviewed.

Introduction

The catalytic oxidation of volatile substances such as methanol and ammonia over platinum was observed in the nineteenth century, and has become a major industrial process during the twentieth century. The subject of catalytic oxidation has been thoroughly reviewed by various authors including Golodets [1], Prasad [2], and Margolis [3]. Most of the work reviewed is that of industrial interest. This work falls into roughly two categories: (1) partial oxidation for the manufacture of petrochemicals such as ethylene oxide, maleic anhydride, and acetylaldehyde; and (2) deep oxidation of contaminants in air for emissions control such as found in automotive catalysts. A recent review by Spivey [4] covers the application of catalytic oxidation to dilute organic contaminant streams. There have been no papers to date specifically concerned with control of emissions from air stripping units, and only two actual applications of this technology in the field have been documented.

Catalytic destruction of such emissions differs from other catalytic pollution control systems in several respects. First, the concentration of the contaminants in the gas phase is quite low, in most cases less than 100 ppmv. Second, the humidity levels are unusually high since the air stream is saturated coming off the stripping unit. Third, a wide variety of contaminants and mixtures ranging from chlorinated compounds and soluble fuel fractions to pesticides and polychlorinated biphenyls may be in any groundwater source. Finally, the water vapor stream exiting a groundwater stripper may have high concentrations of mineral aerosols and poisons (e.g., hydrogen sulfide) that will shorten catalyst life. All of these factors make the design and implementation of catalytic treatment uncertain. Some of these uncertainties are to be considered here, in addition to industrial resources and system economics.

Fundamentals for System Design

The majority of catalytic oxidation processes have been developed for the production of oxidized chemical feedstocks such as sulfuric acid, maleic anhydride, and ethylene oxide. In the case of petrochemical feedstocks, the oxidation process is carefully controlled to prevent the reaction from producing the most thermodynamically favored products, carbon dioxide and water.

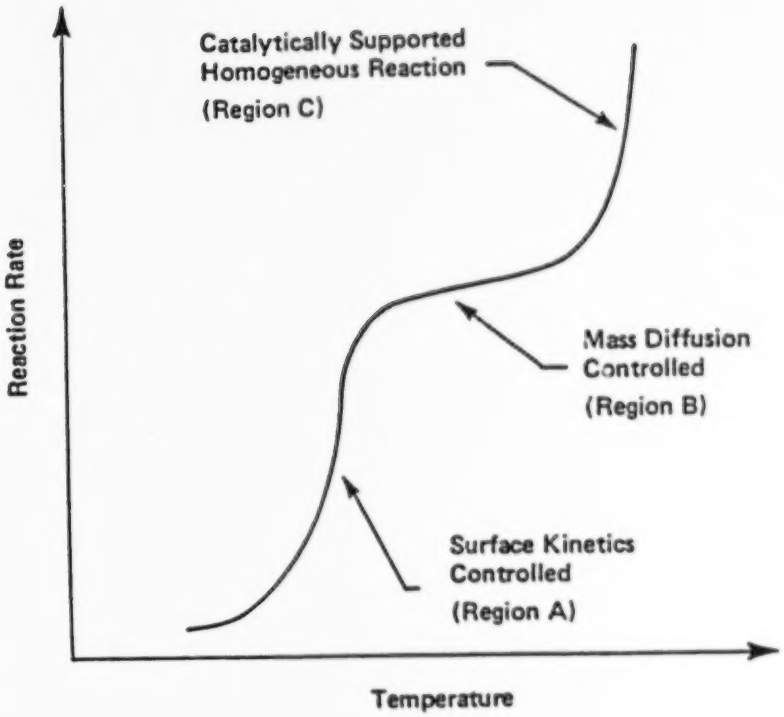
Only in a few cases, most notably automobile emission control systems, is the process designed to promote complete (deep) oxidation of the reactants. In addition, the concentration of reactants in industrial catalytic processes is usually in the percent range; whereas, for contaminant control, the reactant concentrations are very low, usually less than 100 ppmv (0.01%). More specifically, the emissions from air stripping units may consist of a complex mixture of reactants including both fuel and solvent fractions. A typical example of an air stripping off gas stream is shown in Table 1. This table lists the concentration of selected contaminants emitted from a pilot air stripping unit at Wurtsmith Air Force Base in Michigan (Stallings et al. [5]). Here, no individual contaminant exceeds 70 ppmv, but the cumulative total exceeds 170 ppmv. These concentrations are in the range of those seen by automotive catalysts; however, automotive catalyst units cannot be directly compared, because they are also designed to simultaneously accomplish the oxidation of carbon monoxide and the reduction of oxides of nitrogen.

Even though the oxidation reactions in both cases are highly exothermic, the concentrations of reactants from an air stripper are far too low to supply the heat necessary to raise the catalyst bed temperature. Usually, this temperature must be high enough to provide a practical reaction rate for destruction. Figure 1 shows the effect of catalyst operating temperature on the rate of oxidation. The rate of oxidation initially increases exponentially with temperature, and then levels off as the rate limiting of reactants to the catalyst surface becomes the rate limiting factor. At higher temperatures yet, the reaction no longer relies solely on the surface reaction, and the region of catalytic supported combustion dominates. In the case of automotive catalysts, the exit gases from the engine are sufficiently hot to provide this energy, but the air from stripping units is usually near groundwater temperatures, approximately 5° to 25°C. This is far below the operating temperatures of available oxidation catalysts; therefore, heat must be provided in the form of an electric heater or fuel fired burner. To promote energy savings, one would ideally desire the temperatures to be the minimum at which the desired destruction efficiency can be accomplished. This usually corresponds to the mass diffusion controlled regime in Figure 1.

TABLE 1. TYPICAL ORGANIC CONCENTRATIONS IN AIR STRIPPING OVERHEAD [5]

Compound	Concentration in gas phase (ppmv)[a]
Pentane	42.4
Cyclohexane	66.1
Methyl cyclopentane	7.7
2,3-Dimethyl butane	1.6
Trichloroethylene	17.9
Benzene	9.2
Ethyl benzene	8.3
Cumene	3.4
Mixed Xylenes	21.3
Total	177.9

[a]Calculated at a gas/liquid ratio of 42/1 for 1-in. Pall rings.



Source: Prasad et al. [2].

Figure 1. Reaction regimes for catalytic oxidation.

The energy burden of heating a cold, water-laden air stripping effluent stream should be lower for a catalyst system than for thermal incineration, but it still represents a considerable operating cost. The air from most air stripping units can be considered to be saturated with water (100% relative humidity). This corresponds to an absolute water vapor concentration of between 0.8 and 3.1% in the exit stream. The action of bubbles in the stripping unit also produces a large amount of liquid aerosol. The carryover of liquid water in the form of small droplets and aerosol represents an additional large heat burden on any catalyst bed and should be minimized by the use of demisters or filters if possible. Aerosol removal has an additional advantage, in that mineral or biological matter which might foul and deactivate the catalyst bed is also removed.

The high concentration of water vapor in the effluent stream represents a major difference between this application of catalytic control and others which have been previously investigated. Hence, the effect of high humidity on the catalytic chemistry of hydrocarbon oxidation and the destruction of other contaminants has not been previously ascertained. The oxidation of hydrocarbons is generally thought to occur via a surface redox cycle known as the Mars-van Krevelan mechanism (Golodets [1]). This mechanism results in a rate expression of the form:

$$r = \frac{k_{O_2} k_{HC} C_{O_2} C_{HC}}{k_{O_2} C_{O_2} + \gamma k_{HC} C_{HC}}$$

Here k_{O_2} is the rate constant for catalyst surface oxidation, k_{HC} is the rate constant at which the hydrocarbon reduces the oxidized surface, C_{O_2} and C_{HC} are the respective concentrations of oxygen and hydrocarbon, and γ is the stoichiometric requirement of oxygen necessary to completely convert the hydrocarbon to water and carbon dioxide. A multicomponent form of this equation has also been developed by Gangwal et al. [6]. The products of reaction do not appear in this rate expression, but Golodets [1] has postulated that water and carbon dioxide can inhibit the reaction by competitively adsorbing on reaction sites. Preliminary experimental studies by the authors on the effect of humidity on catalytic oxidation of hydrocarbons indicate that this

inhibition effect is minimal for most hydrocarbons in fuel fractions. However, for compounds which may hydrolyze (e.g., ethyl acetate), water may actually enhance the oxidation rate. As of now, however, no firm basis exists to predict the behavior of hydrocarbon oxidation in the presence of water.

The effect of water on the catalytic destruction of chlorinated compounds is generally thought to be beneficial. The most easily treated products of this process are hydrochloric acid (HCl), carbon dioxide, and water; however, some solvents such as trichloroethylene (C_2HCl_3) do not have stoichiometric amounts of hydrogen to allow complete reaction of the chlorine to HCl. For those compounds, water may serve as this hydrogen source, or may merely favorably alter the reaction thermodynamics. Other hydrogen sources such as methane or propane may also be beneficial in promoting HCl formation. As with the non-halogenated hydrocarbons, a clearer understanding of the catalytic chemistry is essential to understanding the role of water vapor.

As a further complication, many contaminated groundwater sources may contain mixtures of hydrocarbons and halogenated compounds. Gangwal [6] and Tichenor [7] have found significant mixture effects for oxidation of hydrocarbons, chiefly in the form of inhibition. This means that catalytic oxidation of a given compound is generally negatively impacted by the presence of mixtures, and higher catalyst bed operating temperatures may be necessary to achieve adequate destruction. To date, information on mixture effects for halogenated compounds is extremely limited, although Doronina [8] and others in the Russian literature report successful catalytic treatment of organic and halogenated compound mixtures.

Catalysts exposed to air stripping effluent are subject to deactivation from several sources - fouling, thermal degradation, poisoning, and catalyst volatilization. Due to dissolved salts and mineral matter in the off gases from the stripping operation, the catalyst bed may plug or become encrusted and lose effectiveness. As mentioned previously, removal of aerosols prior to the catalyst unit may alleviate this problem. If the catalyst bed is operated at high temperatures (>600°C) for prolonged periods, sintering of active noble metals may occur also resulting in loss of activity. Groundwater also often contains significant amounts of readily strippable compounds, such as hydrogen sulfide (H_2S), that may poison the catalyst surface by adsorbing and blocking

active sites. Higher operating temperatures may remove some of these poisons, but sintering may then become a problem. Finally, some reactants, especially halogenated compounds, may chemically bond with the active catalyst and subsequently volatilize from the surface. The loss of catalyst associated with this vaporization process inevitably results in lower catalyst efficiency.

Catalytic Units for Contaminant Removal

Ideally, to accomplish the catalytic deep oxidation of contaminants, a highly active, non-selective catalyst is desired. The compositions of commercial catalysts for deep oxidation are generally proprietary in nature, but generally consist of low weight loadings of platinum or palladium (0.1 to 3%) on metal or metal oxide substrates. Sometimes the noble metals are supplemented by a base metal component or a metal oxide wash coat on the support. For application to halogenated compounds, metal oxides of compounds such as copper, vanadium, or chromium may show superior resistance to deactivation. The large body of literature on the formulation and chemistry of oxidation catalysts cannot be adequately summarized here, but reviews by Golodets [1], Spivey [4], and others provide an introduction to the subject.

A catalyst with high activity at low temperatures is desirable, but enough heat must be supplied to effect complete destruction. However, extremely high temperatures (>750°C) may actually produce more partial reaction products. Basically, this means that a minimum operating temperature of at least 350°C should be employed with catalysts currently available. To attain these temperatures, most catalyst units, regardless of the catalyst bed structure, use a preheating unit. This preheater may be electric or an open flame type. The open flame type is generally powered by natural gas or propane, and may accomplish a significant degree of destruction by itself (Tichenor [7]). These preheaters are used with most of the reactors now available commercially, in both fixed and fluidized bed designs.

There are several commercial catalytic units now on the market in this country, and many others in Europe. To our knowledge, however, there is experience with only two of these in conjunction with an air stripping unit. Most of these units are now used for controlling emissions from solvent stripping operations, paint spray booths, and similar industrial operations where concentrations of reactants are much higher. Several of the commercially available catalytic incinerators have been identified and are described below:

1. ARI: This fluidized bed catalytic incinerator was a new development in the early 1970s. Developed jointly by Air Resources, Inc. and Harshaw Chemical Company, this technology has been in commercial use since 1975 and is being marketed on a turnkey basis (Hardison [9]). The catalyst, developed by Harshaw Chemical Company, is a nonprecious metal disposable pellet. The fluidized design is especially good in that it overcomes carbon fouling (masking) problems. The moving catalyst bed allows masked catalyst to move to higher temperature zones in the bed, thereby oxidizing the masking material. About 15 of these fluidized bed units are being used for volatile organic compound (VOC) control (1981). Of the 15, a significant portion are used for control of chlorinated hydrocarbons.

2. Torvex: Since the early 1980s, this system has been marketed by the Engelhard Corporation. Complete systems are available in sizes ranging from less than 1,700 to 120,000 m^3/hr. Their catalysts consist of precious metal deposited on ceramic honeycombs. Between 1976 and 1981, the vendor supplied about 100 complete catalytic incinerator systems and retrofitted about 200 thermal incinerators with catalyst.

3. Ultrox: Ultrox International continues to develop and market a system based on a combined ultraviolet (UV)/ozone/catalytic oxidation technology. When used for water treatment, the groundwater contaminated with chlorinated hydrocarbons is pumped into a reaction tank where an ozone/air mixture is bubbled through the water. This bath is simultaneously irradiated with UV light. In addition to the oxidation in the aqueous phase, a portion of the hydrocarbons are stripped from the water and pass into a UV/catalytic reactor where further oxidation occurs at temperatures slightly above ambient.

4. Groundwater Technologies: Groundwater Technologies provides complete groundwater cleanup services. As such, they have entered the area of cleanup of air emissions from these operations. They report installation of their first catalytic conversion unit. The catalyst was obtained from Johnson-Matthey and consists of platinum on a metal substrate. Effective destruction of fuel mixtures stripped from groundwater has been cited [10].

5. CE Air Preheater: CE markets both thermal and catalytic incinerator systems in sizes ranging from 3,400 to over 170,000 m^3/hr. Their catalyst, also supplied by Johnson-Matthey, is platinum deposited on a stainless steel honeycomb. CE Air Preheater had installed approximately 10 catalytic incinerator systems as of 1982.

6. Met-Pro Corporation: Formerly Oxy-Catalyst, this company markets both thermal and catalytic incineration systems and carbon adsorption systems. They employ a wide range of catalysts including both precious and nonprecious metals deposited on a monolithic or pellet support. They have installed over 200 catalytic incinerator systems throughout the United States and some foreign countries (Kenson [11]).

7. TEC Systems: This producer manufactures and markets catalytic incinerator systems that utilize a granular catalyst bed. These incinerators have been effectively used to control VOC emissions from a number of industrial processes.

8. Dedert Corporation: Dedert markets a "CATOX" catalytic incinerator for Haldor Topsoe A/S, a Danish company specializing in catalysts. It features a non-noble metal catalyst resistant to chlorine, sulfur, and phosphorus compounds in both monolith and pellet form. They market a special ADOX unit for low concentrations (<250 ppmv), and another, named REGENOX, for "dirty" streams. Many of these units are in place for manufacturing and industrial proces es.

Case Studies

The U. S. Environmental Protection Agency (EPA), during the past 3 years, has sponsored or co-sponsored several evaluations of catalytic control devices. During 1984, a Torvex unit from Engelhard Corporation was evaluated using low concentrations of several compounds (Tichenor [7]). The objectives were to investigate the effects of operating and design variables on destruction efficiency in mixtures of contaminants. Tests showed that destruction efficiencies exceeding 95% could be attained for alcohols, acetates, ketones, aldehydes, aromatics, and ethylene oxide. Chlorinated compounds were not effectively destroyed with this catalyst.

The EPA and the U.S. Air Force Engineering and Services Center cooperated to evaluate other off-the-shelf technologies including the fluidized bed unit from ARI and the UV/ozone/catalytic unit from Ultrox (Palazzolo et al. [12]). For the Ultrox unit, the destruction of chlorinated compound mixtures was evaluated with both ozone addition and UV light. It was found that destruction efficiencies without ozone addition in the presence of UV light were low. The presence or absence of UV light with ozone addition did not have a significant effect on destruction efficiency, which was 99% in either case. However, unidentified by-products were actually being formed in the presence of ozone which lowered overall destruction efficiency to approximately 75%.

A recent EPA report [13] identified 177 air strippers in the United States. Of these, 17 are equipped with air emission controls. Of the 17 facilities, 1 uses a catalytic incinerator, 2 have open flares, 2 have thermal incinerators, and 12 have granular-activated carbon adsorbers. Installation of the air control device was required by the governing state at nine of these sites.

The catalytic incinerator identified in that study is located at the U.S. Coast Guard Base in Traverse City, Michigan. This air stripper system began operation in 1985. The catalytic incinerator was included in the initial design and installation of the air stripping system. The air stripper installed at this site is a rotary high-gravity air stripper which achieves high removal efficiencies at lower air to water ratios than packed towers. The lower air flow for this type of air stripper results in higher pollutant concentrations in the air stripper exhaust. The catalytic incinerator at Traverse City was installed for destruction of benzene, toluene, and xylene stripped from the groundwater. The catalytic oxidation unit is designed for a flow of 3,400 m^3/hr and operates at 260° to 320°C. The design efficiency for this catalytic incinerator was 90%; however, no performance data are yet available.

The performance of catalytic incinerators has been demonstrated for control of organic air emissions from air stripping of groundwater in one other case [13]. The destruction performance achieved during pilot scale testing of an air stripper exhaust stream containing benzene and toluene is presented graphically in Figure 2. The incoming air from the stripping unit contained less than 10 ppm total organics. It can be readily perceived from the lack of performance data on field systems that a large portion of research in this area remains to be conducted.

System Economics

Two recent studies have evaluated the cost of control technologies for air emissions from groundwater stripping. The first of these was completed for EPA's Control Technology Center in August 1987 [13]. Here the costs for the Traverse City site are estimated. Installed cost for the catalytic unit is $198,500, and annual operating costs for the 3,400 m^3/hr unit is $95,920 (third quarter 1986). This report also estimates control costs and efficiencies for carbon adsorption and thermal incineration for four other air

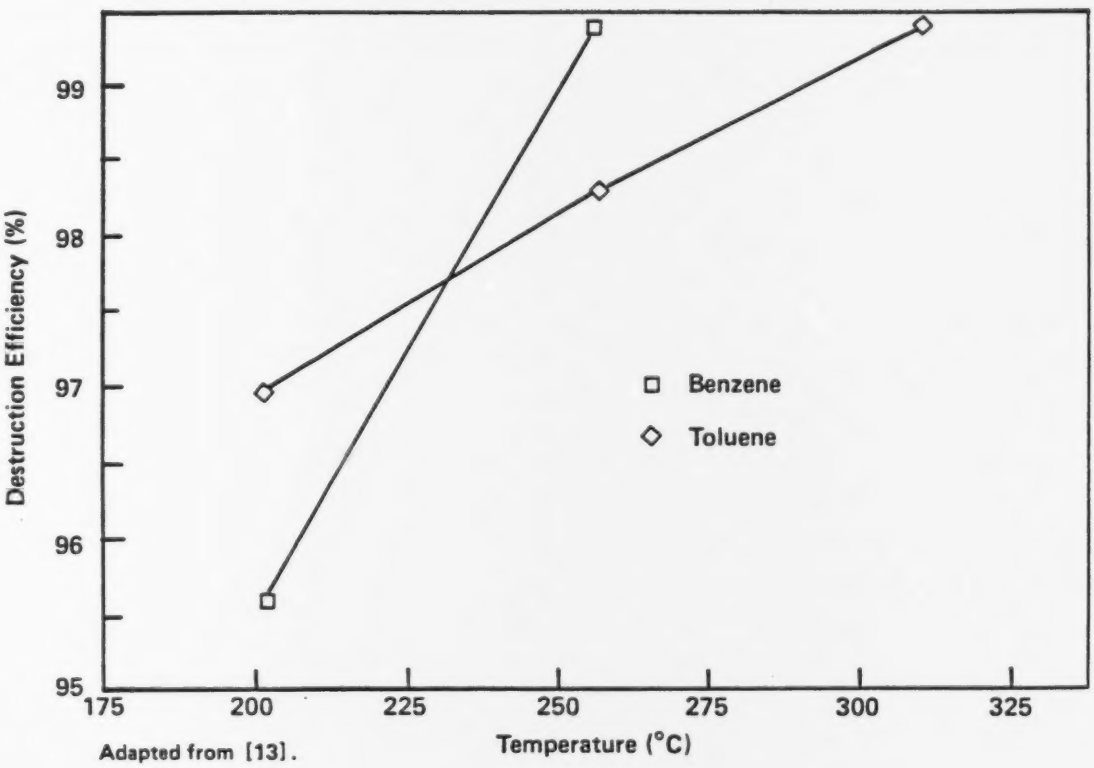


Figure 2. Destruction efficiency of a pilot scale catalytic incinerator for benzene and toluene in air stripper exhaust.

stripper sites. Table 2 summarizes these estimates. It may be noticed from this table that the annualized costs of control increase as a function of air flow rate for all of the systems. The most rapid increase in cost is for thermal incineration, with catalytic oxidation being twice as cost effective for the 32,600 m^3/hr unit. This emphasizes the energy savings mentioned previously for catalytic systems. Carbon adsorption still has a more favorable operating cost; however, the control efficiency is much higher for thermal and catalytic incineration (>95%) than it is for adsorption (70 to 90%). The tradeoffs between control efficiency and costs must be made in light of the nature of the contamination threat.

In a second report, the U.S. Army Toxic and Hazardous Materials Agency (USATHAMA) recently published a study comparing the economic viability of several control technologies for air emissions from groundwater stripping and soil purging operations [14]. The objective of this study was to identify those emerging technologies which could be competitive with traditional controls. Among the options considered were the fluidized bed incinerator of ARI, the UV/Ozone/Catalytic Oxidation unit from ULTROX, the PURASIV fluidized carbon adsorption system of Union Carbide, and the KPR/incineration system marketed in the United States by Met-Pro. This last unit uses an activated carbon fiber adsorbent and special equipment to preconcentrate contaminant fumes prior to incineration. For 1 year of operation for a 1,400 to 12,000 m^3/hr unit, catalytic oxidation and carbon adsorption were the most cost competitive. UV/ozone and thermal incineration were less cost effective at higher flowrates; and the PURASIV and KPR processes were almost twice as expensive per year than standard carbon adsorption at all flowrates examined. The final recommendation of the report was to pursue a demonstration of a fluidized bed catalytic oxidation unit.

Conclusion

Catalytic destruction of groundwater air stripping emissions is an attractive technology in that: (1) the contaminants are destroyed, not simply transferred to another phase; (2) destruction efficiencies in excess of 95% are readily attained; (3) it represents a cost effective alternative to other technologies available; and (4) commercial units are currently available for use. Unfortunately, aside from one limited study, there exists no

TABLE 2: SUMMARY OF AIR STRIPPING EXHAUST CONTROL SYSTEMS [13]

	Site A	Site B	Verona Well Field	Plainfield
Contaminants Concentration (ppbw)	Trichloroethylene (4,000) Trichloroethane (300)	Chloroform (1500)	Ethylene dichloride (5) Trichloroethane (12) Dichloroethylene (10) Trichloroethylene (1) Perchloroethylene (10)	Perchloroethylene (200)
Water Flowrate (lpm)	5,300	590	7,200	13,600
Air Flowrate (st. m^3/hr)	13,600	2,200	9,400	32,600
Organic Removal Efficiency (%)	99	99.9	99.9	99.6
Control Options:				
Carbon Adsorption				
Control efficiency (%)	~80	not reported	74	90
Installed cost ($)	150,000	152,000	223,000	500,000
Direct operating cost ($)	50,400	31,300	62,700	120,000
Total annualized cost ($)	96,400	77,800	124,000	221,400
Thermal Incineration				
Control efficiency (%)	98	98	98	98
Installed cost ($)	318,000	187,000	285,000	432,000
Direct operating cost ($)	432,000	86,800	303,000	1,047,000
Total annualized cost ($)	492,000	129,000	358,000	1,123,000
Catalytic oxidation				
Control efficiency (%)	95	95	95	95
Installed cost ($)	307,000	135,000	251,000	586,000
Direct operating cost ($)	210,000	51,600	150,000	508,000
Total annualized cost ($)	269,000	86,100	201,000	606,000

experimental database on which to quantitatively evaluate this approach. Control of emissions from air stripping units differs from other similar applications because of the low concentrations of contaminants, the high humidity, and catalyst deactivation mechanisms. Laboratory scale experiments to understand the chemistry of reaction for different contaminant types and to evaluate catalyst formulations and activities will play an important role in removing technical uncertainties. Matching of catalyst to contaminant, selection of reactor type and operating temperatures, and development of techniques to realistically evaluate performance will greatly aid transfer of the technology to the field.

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